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# Was the Himalayan orogen a climatically significant coupled source and sink for atmospheric CO<sub>2</sub> during the Cenozoic?

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#### **Abstract**

The hypothesis that the Himalayan orogen was a climatically significant coupled source and sink for atmospheric  $CO_2$  during the Cenozoic is evaluated in light of the timing, duration and  $CO_2$  fluxes associated with Himalayan metamorphism and chemical weathering. We suggest that diachronous Eohimalayan metamorphism occurred over a  $\sim$ 20 m.y. time span (Middle Eocene to Early Oligocene) with total metamorphic  $CO_2$  production of  $\sim$ 4–10 × 10<sup>18</sup> mol. Because this is much greater than the amount of carbon stored in the atmosphere and oceans, and because uplift and accelerated erosion began at least  $\sim$ 5 m.y. after the peak of metamorphism, we conclude that it is implausible that  $CO_2$  produced by metamorphism in the Himalayan orogen was consumed millions of years later by erosion-enhanced weathering in this orogen. Assuming a global climate/silicate-weathering feedback, we estimate that metamorphic  $CO_2$  degassing from the Himalayan orogen would have produced a warming of <0.5°C, and enhanced weathering in this orogen would have produced a cooling of <0.2°C; thus, direct climate effects of this degassing and weathering were likely to have been minor. © 1999 Elsevier Science B.V. All rights reserved.

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#### 1. Introduction

Bickle [1,2] attributed the 50–100 m.y. episodicity in the seawater <sup>87</sup>Sr/<sup>86</sup>Sr curve to the episodic nature of collisional orogenesis. He hypothesized that, for each cycle, CO<sub>2</sub> added to the atmosphere from post-collisional regional metamorphism was consumed by enhanced chemical weathering accompanying subsequent uplift and erosion.

For the Himalayan orogen Bickle [1] concluded

that a 50% linear increase in  $CO_2$  drawdown by silicate weathering over the past 40 m.y. consumed  $13-26 \times 10^{18}$  mol of  $CO_2$ . His computed metamorphic  $CO_2$  release of  $24-60 \times 10^{18}$  mol during Himalayan regional metamorphism more than compensated for the consumption of atmospheric  $CO_2$  by chemical weathering.

Bickle [1,2] concluded that imbalances of  $\pm 25\%$  in the CO<sub>2</sub> degassing vs. weathering fluxes were implied by the seawater Sr-isotope curve. However, quantitative modeling of the global carbon cycle [3–6] illustrates that CO<sub>2</sub> degassing and weathering fluxes for time periods > 1 m.y. must be well within

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 $\pm 25\%$ . Thus, for a silicate weathering CO<sub>2</sub> flux of  $8 \times 10^{18}$  mol m.y.<sup>-1</sup> [6], imbalances between Earth degassing fluxes and silicate-rock weathering fluxes must have been  $< 2 \times 10^{18}$  mol m.y.<sup>-1</sup> (cf. Bickle [1]).

Here we assess the validity of the hypothesis that CO<sub>2</sub> added to the atmosphere by Himalayan metamorphic degassing was consumed by chemical weathering in this orogen. We quantify imbalances in CO<sub>2</sub> sources and sinks associated with the Himalayan orogen through estimates of the timing and rates of metamorphic CO<sub>2</sub> degassing and CO<sub>2</sub> drawdown by silicate weathering, and to compare these imbalances with estimates of maximum plausible imbalances inferred from paleoatmospheric CO<sub>2</sub> levels during the Cenozoic.

### 2. Constraints on maximum Cenozoic atmospheric CO<sub>2</sub> content

Freeman and Hayes [7] concluded that the maximum atmospheric  $CO_2$  contents in the Cenozoic were about twice the modern value, with significant uncertainties [5,8]. Maximum atmospheric  $CO_2$  contents of 1300 ppm have been estimated for the Late Cretaceous [9]. Because the atmospheric  $CO_2$  levels of the Cenozoic were lower than the Cretaceous [7,10], we adopt  $\sim 1000$  ppm as an upper limit for Cenozoic atmospheric  $CO_2$  concentration.

#### 3. Computational strategy

We constrain maximum values of the imbalance in atmospheric CO<sub>2</sub> input vs. output consistent with estimated Cenozoic atmospheric CO<sub>2</sub> levels. We use a simple model of ocean chemistry and air—sea gas fluxes [6] to estimate the changes in atmospheric CO<sub>2</sub> content resulting from inferred CO<sub>2</sub> fluxes associated with the Himalayan orogen, in the absence of other negative stabilizing feedbacks, such as the climate/silicate-weathering feedbacks, such as the climate/silicate-weathering feedback [3,4,6,11]. If, as we demonstrate, the inferred change of atmospheric CO<sub>2</sub> content resulting from Himalaya-related CO<sub>2</sub> fluxes are implausible, this strongly suggests that the proposed coupling between metamorphic degassing and orogenically induced weathering within the Himalayan orogen was insufficient to maintain

global balance in the carbon cycle, and that global climate/silicate-weathering feedback maintained a near-balance between atmospheric inputs and outputs of CO<sub>2</sub>.

### 3.1. Quantifying effect of degassing/weathering imbalances

To quantify the effect of imbalances between CO<sub>2</sub> degassing and silicate-rock weathering on ocean chemistry and atmospheric  $pCO_2$  we used the ocean-atmosphere model of Berner and Caldeira [6]. The essential assumptions are: the ocean consists of deep and surface reservoirs of carbon and alkalinity, the atmosphere is a single reservoir of carbon; the surface ocean is in equilibrium with atmospheric CO<sub>2</sub>; and the deep-ocean is in equilibrium with calcite. Our basic computational approach is to add carbon from excess degassing or alkalinity from excess weathering and let (1) CO<sub>2</sub> in the atmosphere equilibrate with the surface ocean, (2) the carbon and alkalinity in the surface ocean equilibrate with that of the deep-ocean, (3) carbon and alkalinity in the deep-ocean equilibrate with calcite in marine sediments, and (4) the surface- to deep-ocean gradients of total dissolved inorganic carbon and total alkalinity remain constant. In the case of excess degassing, equilibration with sediments involves carbonate dissolution, whereas with excess weathering, equilibration with sediments involves carbonate precipitation. As this equilibration process approaches steady state, we track atmospheric  $pCO_2$ .

Carbonate chemistry was computed using the equations and rate constants described in Stumm and Morgan [12], assuming a deep-ocean temperature of 2°C and the surface ocean at a temperature of 18°C and with a salinity of 35‰. The carbonate-ion concentration of the deep-ocean is a function of deep-ocean total dissolved inorganic carbon and alkalinity:

$$\left[ \text{CO}_3^{2-} \right]_{\text{deep}} = \text{CO}_3 \left( \left[ \sum \text{CO}_2 \right]_{\text{deep}}, \left[ \sum \text{Alk} \right]_{\text{deep}} \right)$$

The system is solved assuming  $[CO_3^{2-}]_{deep} = 90$   $\mu$ mol kg<sup>-1</sup>. Surface total dissolved inorganic carbon and alkalinity concentrations differ from the deep-ocean values due to both marine biology and chemical solubility [13]. Here, we consider the dif-

ference between surface values and deep value to be constant:

$$\left[\sum CO_{2}\right]_{surface} = \left[\sum CO_{2}\right]_{deep} - \Delta \left[\sum CO_{2}\right] (2)$$

and

$$\left[\sum Alk\right]_{surface} = \left[\sum Alk\right]_{deep} - \Delta \left[\sum Alk\right] \quad (3)$$

where  $\Delta[\sum CO_2] = 290 \,\mu\text{mol kg}^{-1}$  and  $\Delta[\sum Alk] = 92 \,\mu\text{eq kg}^{-1}$  [13]. Surface  $pCO_2$  is computed based on surface total dissolved carbon and alkalinity concentrations as per Stumm and Morgan [12]:

pCO<sub>2, surface</sub>

= 
$$p \text{CO2} \left( \left[ \sum \text{CO}_2 \right]_{\text{surface}}, \left[ \sum \text{Alk} \right]_{\text{surface}} \right)_{(4)}$$

The simplified model is intended to represent gross features of ocean chemistry that are important to the problem at hand. Results would be qualitatively similar with other reasonable assumptions. For example, the deep-ocean carbonate-ion concentration could have been determined by a flux constraint on alkalinity instead of making the assumption of constant carbonate-ion concentration. Over the course of the Cenozoic, the calcite compensation depth (CCD) was no more than  $\sim 1$  km shallower than today [14]. At constant temperature and calcium-ion concentration, a 1 km shallowing of the calcite lysocline would imply a  $\sim 15\%$  reduction in the carbonate-ion concentration [15]. Using the equations in Stumm and Morgan [12], we compute that at constant pH, this change in the CCD depth would correspond to a proportional reduction in atmospheric CO<sub>2</sub> content. In contrast, at constant total alkalinity this CCD depth change would imply a  $\sim$ 25% increase in atmospheric CO<sub>2</sub> content. Uncertainties of this magnitude would not affect our conclusions.

## 3.2. Quantifying effect of changes in atmospheric $CO_2$ from degassing and weathering

To quantify the effect of imbalances in CO<sub>2</sub> degassing and weathering on atmospheric CO<sub>2</sub> content, we apply the computed amount of excess CO<sub>2</sub> (or alkalinity, in the case of excess weathering) to the model described above and in [6]. Sundquist [3] provided an analysis of the global response to 10%

imbalances in fluxes of  $CO_2$  degassing vs.  $CO_2$  drawdown by weathering. He found that if the silicate-weathering feedback acted as has been proposed, that the time-scale to approach a new steady-state atmospheric  $CO_2$  content is  $\sim 300,000-400,000$  year.

#### 4. Himalayan metamorphism

#### 4.1. Timing and duration of metamorphism

Analysis of the timing and duration of Himalayan metamorphism is necessary to estimate the rate of CO<sub>2</sub> degassing and to assess the temporal link between CO<sub>2</sub> degassing and atmospheric CO<sub>2</sub> drawdown by chemical weathering. Bickle [1] proposed such a temporal link, but did not carry out an assessment of the timing of the degassing or the plausibility of the time lag between enhanced degassing and enhanced weathering.

Cenozoic regional metamorphism in the Himalaya is manifest in the High Himalayan Crystalline Series (HHCS). Radiometric dating [16–18] of the HHCS in central Nepal suggests two periods of metamorphism: Barrovian-type prograde Eohimalayan metamorphism followed by retrograde Neohimalayan metamorphism.

#### 4.1.1. Eohimalayan prograde metamorphism

Timing of the initiation of prograde metamorphism is constrained by the age of collision. Due to counterclockwise rotation of the Indian plate, collision in the Himalayan orogen was diachronous: ~52 Ma in the west and ~41 Ma in the east [19]. Diachroneity of the India—Asia collision would have resulted in a corresponding eastward propagation of post-collisional metamorphism in the Himalayan belt. Based on geochronologic data Guillot et al. [18] concluded that the Eohimalayan metamorphism occurred in the Mid-Eocene to Early Oligocene in the western part of the orogen and Late Oligocene in the eastern part. However, recent geochronologic data suggest that the prograde metamorphism in the western part of the belt could have continued into the Late Oligocene [20].

#### 4.1.2. Neohimalayan retrograde metamorphism

In the central Himalaya (Nepal), Neohimalayan metamorphism occurred from Late Oligocene through Middle Miocene [16,17]. The Neohimalayan event was accompanied by brittle deformation and fluid circulation along the Main Central Thrust [16]. Widespread shallow metamorphic CO<sub>2</sub> degassing *could* have accompanied extensional tectonism throughout the Neohimalayan metamorphism [11]. As with the Eohimalayan metamorphism, diachronous collision would have resulted in the eastward propagation of the Neohimalayan metamorphism.

#### 4.2. Metamorphic CO<sub>2</sub> degassing

To compute the total CO2 evolved during Himalayan metamorphism, Bickle [1] inferred a value of  $5 \times 10^7$  km<sup>3</sup> for the total volume of rock that underwent metamorphism from the volume and composition of sediment in the Himalayan foreland basins and submarine fans [21]. He derived the amount of CO<sub>2</sub> released by metamorphism in two ways: (1)  $\sim 60 \times 10^{18}$  mol, computed by arbitrarily assuming that the protolith contained 5 wt% carbonate and implicitly assuming that this carbonate was completely consumed by metamorphic decarbonation (termed the 'mass loss' method [11]), and (2)  $24 \times 10^{18}$  mol, computed using an average fluid flux during metamorphism of 10<sup>3</sup> m<sup>3</sup> m<sup>-2</sup>, arbitrarily assuming a 10 km path length for fluid flow, and an estimated fluid composition of 10 mol% CO<sub>2</sub>.

Johnson's [22] more conservative estimate of eroded volume ( $2 \times 10^7 \text{ km}^3$ ) would reduce Bickle's total estimated CO<sub>2</sub> production from 24 to  $60 \times 10^{18}$  to 10 to  $24 \times 10^{18}$  mol. Bickle's computations implicitly assume that the sediment contained in the foreland basins and the Indus and Ganges–Brahmaputra fans was entirely derived from the HHCS; however, up to 20% of the Bengal Fan detritus in the past 20 m.y. may have been derived from erosion of other units [23]. This factor would further reduce Bickle's [1] estimated CO<sub>2</sub> loss to 8 to  $19 \times 10^{18}$  mol.

Rather than using the volume of sediment in the Himalayan foreland basins and Bengal fans, we derived a more conservative volume of rock undergoing Himalayan metamorphism by considering the exposed  $\sim 2.5 \times 10^5 \text{ km}^2$  outcrop area of the HHCS [11]. Assuming that  $\sim 35 \text{ km}$  of overburden was removed by erosion [22], the total volume of HHCS removed would be  $\sim 9 \times 10^6 \text{ km}^3$ . Bickle's

[1] assumption that the Himalayan metamorphism involved mostly pelitic protoliths is supported by the predominance of metapelites in the HHCS of central Nepal [16]. Assuming that the HHCS protolith was a pelite with 5 wt%  $CO_2$  [1] and that all carbonate was consumed during metamorphism [11], the total amount of  $CO_2$  released during metamorphism of the HHCS computed by the mass loss method would be  $\sim 11 \times 10^{18}$  mol. Alternatively, if we apply our estimated  $CO_2$  flux of  $\sim 1.5 \times 10^{12}$  mol km<sup>-2</sup> Ma<sup>-1</sup> [11] to the  $\sim 2.5 \times 10^5$  km<sup>2</sup> exposure of the HHCS, and assume that prograde metamorphism lasted 10 m.y., the total  $CO_2$  flux would be  $\sim 3.75 \times 10^{18}$  mol.

Based on estimating  $CO_2$  evolved by mass loss and by fluid flux data, we conclude that the Himalayan orogen would have generated a total of  $\sim$ 4 to  $11 \times 10^{18}$  mol of  $CO_2$ ; approximately one-sixth the amount estimated by Bickle [1]. Our estimate of  $CO_2$  release is less than that of Bickle because we estimated a smaller volume of rock that underwent metamorphism.

Bickle [1] implicitly assumed that all of the metamorphic CO<sub>2</sub> generated at depth escaped to the Earth's surface. However, the CO<sub>2</sub> content of fluids generated by metamorphism at depth would be diminished by precipitation of carbonate upon cooling and decompression of fluids rising toward the Earth's surface [11,24,25]. Although we cannot quantify the amount of CO<sub>2</sub> consumed by carbonate vein formation during the Himalayan metamorphism, we note that carbonate veins occur in metamorphic rocks of the Nanga Parbat area [26]. Thus, some CO2 was consumed by carbonate vein formation in the Himalayan metamorphic system. As evidenced by massive carbonate formation in major shear zones associated with regional metamorphic belts (such as the Mother Lode system [24]), a significant fraction of deeply generated metamorphic CO<sub>2</sub> could be sequestered upon transport toward the Earth's surface.

Appreciable metamorphic  $CO_2$  could have been released to the atmosphere during extensional tectonism accompanying uplift of the Himalaya [11]. Decarbonation driven by convective circulation of meteoric water in extensional regimes is restricted to the upper  $\sim$ 7 km of the crust above the brittle/ductile transition [11]. If the Neohimalayan event was contemporaneous with rapid erosion, the upper  $\sim$ 7 km of crust undergoing such devolatilization would

have migrated through the underlying HHCS as the Earth's surface was denuded by erosion. Assuming that 35 km of rock was removed from the Himalayan belt by erosion in the past 20 m.y. [22], the average erosion rate would be 1.75 km m.y.<sup>-1</sup>. Coupling this with the assumption that Neohimalayan decarbonation occurred from  $\sim$ 23 Ma (the start of uplift) to  $\sim 13$  Ma (muscovite cooling ages),  $\sim 17.5$  km of the HHCS would have been eroded during this time period. With the exposed surface area of HHCS  $(\sim 2.5 \times 10^5 \text{ km}^2)$  the total volume of HHCS subjected to Neohimalayan metamorphism would be  $\sim$ 4.4  $\times$  10<sup>6</sup> km<sup>3</sup>. If the original protolith had 5 wt% carbonate,  $\sim 5.3 \times 10^{18}$  mol of CO<sub>2</sub> would have been released. However, the CO2 evolved during the Neohimalayan event was likely to have been far less than this because it is probable that most of the carbonate in the HHCS protolith was consumed by decarbonation during the earlier Eohimalayan prograde metamorphism.

### 5. $CO_2$ degassing from Cenozoic Himalayan magmatism

Could Cenozoic magmatism within the Himalaya and Tibetan Plateau have been a climatically significant source of CO<sub>2</sub>? As evidenced by batholiths along the Himalayan orogen, widespread calc-alkaline magmatism occurred prior to collision [27]. During decompression at the onset of rapid exhumation, small volumes of leucogranite formed  $\sim$ 20 Ma. Consequently, it is unlikely that the Himalayan belt degassed significant quantities of magmatic CO2 to the Cenozoic atmosphere. In the Tibetan plateau there was virtually no magmatic activity between 90 Ma and Mid-Miocene [27]. Small volumes of potassic basalts have been erupting in the Tibetan plateau for the past 13 m.y. [27]. Considering the limited solubility of CO<sub>2</sub> in basaltic magmas [28], a climatically insignificant quantity of CO2 would have degassed from the small volume of Neogene potassic volcanics in the Tibetan plateau. We conclude that the Himalayan-Tibetan magmatism was not a significant source of magmatic CO<sub>2</sub> to the Cenozoic atmosphere.

### 6. CO<sub>2</sub> drawdown by Cenozoic Himalayan weathering

Bickle [1] and Tajika [29] computed atmospheric CO<sub>2</sub> drawdown fluxes for the past 40 m.y. based on the assumption that the rise in seawater 87Sr/86Sr ratios during this time period was produced by enhancement of riverine radiogenic Sr contents by chemical weathering of <sup>87</sup>Sr-enriched silicates in the Himalayan metamorphic source rocks. However, coupling the major Sr sources with mass balance computations, the Sr isotopic evolution of the oceans requires that only 20% of the riverine Sr is derived from silicate weathering [30,31]. For the Himalaya orogen this conclusion is supported by Blum et al. [26] who, in a study of a watershed in the HHCS, concluded that 82% of the riverine HCO<sub>3</sub><sup>-</sup> flux is from weathering of carbonate and only 18% from silicate weathering. Even though carbonate composes only a small ( $\sim$ 1%) proportion of the bedrock exposed in this watershed, it has a dominant effect on the dissolved riverine flux. Furthermore, Sr-isotopic compositions of carbonates in paleosols of the Himalayan foreland basin suggest that the marked increase in Himalayan riverine 87Sr/86Sr ratios in the Late Miocene can be attributed to widespread unroofing of highly radiogenic metacarbonate rocks [32]. Based on independent lines of evidence, Quade et al. [32] and Blum et al. [26] concluded that attributing the rise in seawater 87Sr/86Sr ratios over the past 40 m.y. to silicate weathering alone results in overestimating the amount of CO<sub>2</sub> consumed. Accordingly, the increased atmospheric CO<sub>2</sub> consumption in the past 40 m.y. would be significantly lower than that computed by Bickle [1].

Himalayan orogenic detritus first appeared in the Early Miocene [33–35]. Thus, enhanced chemical weathering from uplift of the Himalayan belt would have been confined to the past 23 m.y. rather than the past 40 m.y. as inferred from the Sr-isotopic record. Unroofing of radiogenic carbonates from the Himalayan metamorphic belt in the Early Miocene could have contributed to the apparent increase in the seawater <sup>87</sup>Sr/<sup>86</sup>Sr during the Early Miocene.

Assuming that enhanced  $CO_2$  drawdown by Himalayan weathering began  $\sim\!23$  Ma, an uncompensated weathering-induced Himalayan  $CO_2$  drawdown flux of  $0.064\times10^{12}$  mol yr $^{-1}$  [36] would

have reduced atmospheric  $CO_2$  levels to well below pre-anthropogenic levels (Fig. 2). If Cenozoic atmospheric  $CO_2$  contents were similar to present levels [37], this uncompensated drawdown flux would have depleted atmospheric  $CO_2$  within a few million years.

#### 7. Discussion

If 4 to  $11 \times 10^{18}$  mol of  $CO_2$  were released by diachronous Eohimalayan metamorphism over a 20 m.y. period, and if all of this  $CO_2$  was expelled at a constant rate to the surface, the resulting yearly flux would be  $\sim 0.2$  to  $0.5 \times 10^{12}$  mol. If not compensated by other mechanisms, degassing fluxes  $\epsilon 0.1 \times 10^{12}$  mol yr<sup>-1</sup> would generate atmospheric  $CO_2$  equivalent to the maximum amount estimated for the Cenozoic ( $\sim 1000$  ppm) within  $\sim 5$  m.y. of the beginning of regional metamorphism in the Middle Eocene (Fig. 1). Using Bickle's [1] estimated total  $CO_2$  degassing flux (24 to  $60 \times 10^{18}$  mol), atmospheric

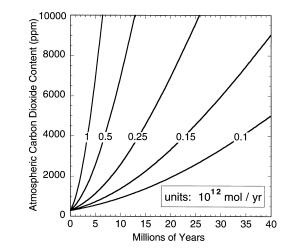


Fig. 1. Change in atmospheric  $CO_2$  concentration (ppm) with selected values of excess  $CO_2$  degassing, barring any compensating feedback mechanisms. The excess  $CO_2$  fluxes are in units of  $10^{12}$  mol yr $^{-1}$ . The initial atmospheric  $CO_2$  content is taken as the pre-industrial value (280 ppm). Excess  $CO_2$  degassing fluxes much greater than  $0.1 \times 10^{12}$  mol yr $^{-1}$  cannot be sustained for many millions of years without generating unrealistic atmospheric  $CO_2$  contents. Therefore, some feedback process, such as increased  $CO_2$  consumption by silicate rock weathering rate with increased global mean temperature, must act to balance the carbon cycle on long time scales [3,4,6].

 $CO_2$  levels of  $\sim 1000$  ppm would have been attained within 1 m.y. from the start of metamorphism. Because major uplift and erosion of the Himalayan orogen began in the Early Miocene (a minimum of  $\sim$ 5 m.y. after the peak of the Eohimalayan prograde metamorphism), the tectonically induced Neogene chemical weathering in the Himalaya could not have been a sink for this CO<sub>2</sub>. Because the Himalayan region was probably an area of subdued topography throughout most of the Oligocene [38], this region would not have been a locus of intense chemical weathering during this period. Consequently, with a climate/silicate-weathering feedback mechanism [4], atmospheric CO<sub>2</sub> generated by Himalayan metamorphism would have been consumed by chemical weathering primarily in areas other than the Himalaya. Using a weathering-rate formulation such as that used by Berner [8], an increase in CO<sub>2</sub> flux of  $0.5 \times 10^{12}$  mol yr<sup>-1</sup> would produce a warming of <0.5°C [25].

 ${
m CO_2}$  drawdown by silicate weathering in the Ganges–Brahmaputra drainage basin is  ${\sim}1.3\%$  of the global total [36]. When scaled to area, this flux is near the world average; thus, the Himalayan system is not an anomalous sink for atmospheric  ${
m CO_2}$  [39]. Nevertheless, greenhouse cooling in the late Cenozoic requires that the rate of atmospheric  ${
m CO_2}$  drawdown by enhanced chemical weathering was slightly larger than the flux of enhanced  ${
m CO_2}$  degassing during this period [40].

Because atmospheric CO<sub>2</sub> drawdown of 0.064 × 10<sup>12</sup> mol yr<sup>-1</sup> from enhanced chemical weathering in the Himalayan system between 23 Ma [39] and the present would have decreased atmospheric pCO<sub>2</sub> to well below pre-anthropogenic levels (Fig. 2), a feedback mechanism that would act to control atmospheric CO<sub>2</sub> content is required. Simultaneous CO<sub>2</sub> drawdown by chemical weathering in other orogenic regions of high relief would have compounded this problem. For example, rapid uplift in the central Andes began  $\sim 20$  Ma [41]. Because  $CO_2$  consumption by chemical weathering in the Andes/Congo system is larger than that of the Himalayan system [36], the combined chemical weathering CO<sub>2</sub> consumption of the Andes/Congo and Himalaya systems is likely to exceed  $0.15 \times 10^{12}$  mol yr<sup>-1</sup>. If not compensated, such fluxes would have consumed atmospheric CO<sub>2</sub> well within 10 m.y. after

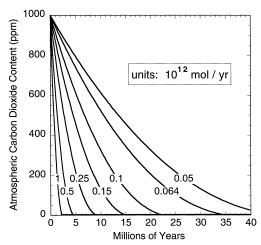


Fig. 2. Change in atmospheric CO<sub>2</sub> concentration (ppm) with selected values of excess atmospheric CO2 drawdown by chemical weathering, in the absence of compensating feedback mechanisms. The excess CO<sub>2</sub> fluxes (inset) are in units of 10<sup>12</sup> mol  $yr^{-1}$ . The curve for  $0.064 \times 10^{12}$  mol  $yr^{-1}$  is based on Galy and France-Lanords [36] estimate of long-term drawdown of atmospheric CO<sub>2</sub> by silicate weathering in the Ganges-Brahmaputra drainage basin. The initial atmospheric CO<sub>2</sub> content (1000 ppm) is the maximum value estimated for the Cenozoic (see text). If uncompensated, CO2 consumption by silicate weathering cannot be sustained for many millions of years without generating unrealistic atmospheric CO2 contents. Therefore, some feedback process, such as diminished CO2 consumption by silicate rock weathering rate with cooler global mean temperatures, or enhanced Earth degassing, must act to balance the carbon cycle on long time scales [3,4,6].

the beginning of the Neogene (Fig. 2). As  $CO_2$  was removed from the atmosphere, the Earth would have cooled and the hydrologic cycle would have decelerated, leading to diminished silicate-rock weathering in other areas [29,42]. Enhanced burial of organic carbon may have been a significant additional sink for atmospheric  $CO_2$  during the Neogene [39]. Using the model described in Kerrick and Caldeira [11] we compute that a regionally enhanced weathering flux of  $0.064 \times 10^{12}$  mol yr<sup>-1</sup> of  $CO_2$  would diminish atmospheric  $CO_2$  content by  $\sim$ 4%, cooling the Earth by  $<0.2^{\circ}C$ . This cooling would occur on the time-scale of 300,000-400,000 year [3], and would be sufficient to lower weathering rates elsewhere to compensate for enhanced Himalayan weathering.

Enhanced magma production at mid-ocean ridges and back-arc basins within the past 10 m.y. [43], coupled with anomalously high CO<sub>2</sub> fluxes from

volcanic [44] and non-volcanic [45] sources, could also have provided additional sources of atmospheric CO<sub>2</sub> during the last half of the Neogene. Increase in the amount of subducted pelagic carbonate throughout the Cenozoic, and consequent expulsion of the CO<sub>2</sub> into the atmosphere by arc volcanism [29,46], could also have provided additional CO2 to the atmosphere. In contrast to a model of removal of atmospheric CO<sub>2</sub> by subduction of carbonate [47], arc magmatism may return of subducted CO<sub>2</sub> to the atmosphere [11]. Furthermore, CO<sub>2</sub> degassing from other magmatic regimes could have had an influence on Cenozoic paleoclimate. For example, the apparent diminution in global temperatures in the early Cenozoic may reflect diminished global magmatic CO<sub>2</sub> expulsion as evidenced by the marked decline in the total volume of mid-ocean ridge and flood basalts during the early Cenozoic [11].

#### 8. Conclusions

Relatively minor imbalances in the rates of input and output of atmospheric CO<sub>2</sub> on time-scales >1 m.y. can produce rather large changes in atmospheric CO<sub>2</sub> content. For example, an imbalance of  $\sim$ 2% between CO<sub>2</sub> sources and sinks, sustained over 10 m.y., would be sufficient to produce unrealistic Cenozoic atmospheric CO<sub>2</sub> contents (Figs. 1 and 2). Metamorphic and magmatic degassing are insufficiently coupled to the tectonic and lithologic factors affecting weathering rates for this link to provide a feedback between CO2 degassing and CO<sub>2</sub> consumption by silicate weathering. Therefore, some feedback must maintain this close balance; this feedback must depend on atmosphere or ocean carbon content [4]. Taking the proposed global climate/silicate-weathering feedback into account [8], relatively minor variations in CO<sub>2</sub> degassing or silicate-rock weathering have a relatively modest impact on global climate.

Our primary conclusions are: (1) because of the long time lag (>5 m.y.) between Eohimalayan metamorphism and enhanced chemical weathering, and the limited capacity of the ocean–atmosphere system to store carbon, the Himalayan orogen was not a coupled source and sink for atmospheric CO<sub>2</sub> during the Cenozoic; (2) in the absence of a negative

feedback, computed Eohimalayan metamorphic CO<sub>2</sub> degassing fluxes would have exceeded the maximum estimated atmospheric CO<sub>2</sub> contents for the Cenozoic within 5 m.y.; (3) without a negative feedback, an estimated atmospheric CO<sub>2</sub> drawdown from enhanced Neogene Himalayan chemical weathering would have produced atmospheric CO<sub>2</sub> contents well below the estimated pre-anthropogenic levels; (4) because these atmospheric CO<sub>2</sub> contents are implausible, a negative feedback mechanism such as the proposed climate/silicate-weathering feedback must function to closely balance atmospheric CO<sub>2</sub> sources and sinks; and (5) due to the global climate/silicate-weathering feedback, CO<sub>2</sub> produced in the Himalayan orogen was likely consumed within a few hundred thousand years by enhanced silicate weathering in other regions of the world.

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